Atmos. Chem. Phys. Discuss., 9, S1407–S1412, 2009
www.atmos-chem-phys-discuss.net/9/S1407/2009/

Atmospheric
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Discussions

Interactive comment on “Modelling the impacts of ammonia emissions reductions on North American air quality” *by* P. A. Makar et al.

Anonymous Referee #1

Received and published: 13 April 2009

This paper is technically sound and addresses an interesting topic. However, the authors present only a limited analysis. The authors examine some topics in great detail, such as a 1% change in the SO_2 dry deposition, but at the end of reading the paper, the cause of the substantial sensitivity of $PM_{2.5}$ and critical load exceedances to NH_3 emissions is not clear. I recommend that the authors extend their analysis to include a more detailed look at these six topics:

1. In Figures 7 and 8, the authors show box and whisker plots of the frequency distribution of the impact of ammonia emissions on $PM_{2.5}$. They note that while the medians of these distributions are close to zero – indicating that the impacts of ammonia emission reductions have only a small impact – there are times when the impacts are much larger. This is an important point. While the annual averaged impact of controlling am-

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monia emissions are likely to be small, there are episodes when the impact is significant. What are the chemical and meteorological conditions that drive these episodes? At the time when the $PM_{2.5}$ is most sensitive to ammonia emissions, does the model accurately represent the nitrate, sulfate and meteorological conditions when compared with the data from the measurement stations? This is critical to building confidence in the results.

2. I strongly agree with the reviewer 2 that the concept of "ammonia-limitation" as described in the introduction is not sufficient for explaining the change in $PM_{2.5}$ due to a change in ammonia emissions. The authors state that "a broader definition of ammonia-limited environments includes a requirement for charge-balancing with nitrate" – such a broader definition is necessary and provided by the work cited by reviewer 2. Figure 3 shows the ammonia-limited regions, and for all seasons California is denoted as not "ammonia-limited". But in Figure 5, California has the largest reduction in $PM_{2.5}$ in the winter, likely due to the change in ammonium nitrate, which is not captured by the "ammonia-limited" metric as defined in this work. In general, the metrics plotted in Figures 3, 9, 10, 11, and 12 are not as helpful to understanding the changes as simply plotting the change in sulfate, nitrate, and ammonium aerosol.

3. The conceptual model in Figure 16 is a useful construct, but not in the way it is currently presented. My sense is that the authors are working to explain why the $PM_{2.5}$ sensitivity in the industrial Midwest of the US and southern Canada is larger than most locations on the continent. This is a valuable goal, but to be useful, some important details are necessary. First, it would be helpful for the authors to more clearly define, perhaps with a map, the regions they are describing. Second, how large is the ammonia mass flux from the eastern emission region to the region of largest $PM_{2.5}$ sensitivity, compared to the emissions in that region of largest $PM_{2.5}$ sensitivity, which are also substantial? It is essential to quantify the fluxes and burdens shown in the figure, before and after the NH_3 emission reduction. These calculations can help the community understand the region of influence of ammonia sources – would it be more

effective to reduce emissions in the region of maximum sensitivity, or upwind as the conceptual model suggests? Finally, the different font sizes are difficult to understand. A simple (+ / $-$) or ($\uparrow$ / $\downarrow$) would be a more clear way to denote change. Some of these may be in error, but it is difficult to check because the differences in font size are subtle. For example, I think that the manuscript explains that that NH_3 dry deposition decreases when emissions decrease, yet the font here is the same size. HNO3 gas dry deposition is missing entirely. However, the authors note a 1% change in SO_2 deposition, and there is a clear font change. I think that the change in NH_3 dry deposition is greater than 1%, but it would be helpful if these values were quantified rather than qualitatively shown in this figure.

4. The authors select a single sensitivity analysis of reducing the Canadian beef cattle emissions by 50%. The impact of this change on $PM_{2.5}$ is very small in magnitude and limited in spatial extent, especially compared to the 30% decrease in all ammonia sources. Furthermore, the locations impacted by this sensitivity test are not areas identified with a critical load. I am puzzled then why the authors conclude that further research on Canadian cattle emissions are of higher priority than other sources. Based on the 30% emission reduction scenario, other sources have greater impact on $PM_{2.5}$ and critical load exceedances. These other sources likely have the same uncertainty and greater impact – what sources contribute most in the regions of greatest $PM_{2.5}$ sensitivity or most sensitive critical load? I realize that these sensitivity tests are time consuming, so I do not require further tests for acceptance of this manuscript. However, the authors should either explain how the results from this test indicate Canadian cattle emission uncertainties are more or less important than uncertainties in other sources.

5. Ammonia emissions are uncertain in their magnitude and timing. For comparison with other work, it would be very helpful to list the magnitude of the ammonia emissions used in this work, divided by month and source category.

6. The change in critical load exceedances in Section 4.2.5 should be explained in more detail. From reading the text and looking at Figure 15, the 30% reduction in NH_3

caused at most a 10% reduction in the magnitude of the exceedance. Is this because most of the exceedance is due to sulfate? The conclusions state that if the ability of the of these ecosystems to absorb nitrogen becomes saturated, then NH_3 emission reductions will become necessary. Does this mean that currently, such emission reductions are not necessary? At current deposition rates, how many years until this changes? Since the change in sulfur due to NH_3 emission reduction is minor, it seems it would make more sense to show the contribution of nitrogen to the exceedance alone and to show the number of years until the capacity to absorb nitrogen is exceeded. From the analysis provided, the impact of changes in nitrogen deposition is not clear.

Specific comments:

P5372@L20: "possibly trans-oceanic consequences downwind" The changes in $PM_{2.5}$ mass in Figure 5 appear to be entirely continental, with no discernable impact offshore. For the deposition changes, there are very strange boundary effects where the deposition changes rapidly go to zero on the eastern boundary, for example, the summer plot in Figure 14. From looking at Figures 5, 13, & 14, I can not see any changes at the boundary. Perhaps it would be possible to calculate the changes at the boundary so the magnitude of the change in trans-oceanic export could be quantified?

P5375@L17-22: Several recent studies have examined the impact of ammonia emission changes on $PM_{2.5}$ specifically for portions of North America (Henze et al., 2008; Pinder et al., 2007, 2008; Tsimpidi et al., 2007). It would be helpful if the authors could report any differences or improvements over this previous work.

P5376@L21: What fraction of the total ammonia / ammonium emissions are as particle ammonium? Does it matter, or are these particles subject to thermodynamic equilibrium immediately after they are emitted? I think this is what implied by the last sentence, but this could be made more clear.

P5379@L13: What are the biogenic emissions of ammonia?

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P5382@L19-26: This manuscript frequently points out that the response to a change in ammonia emissions is non-linear with respect to the concentrations of the inorganic anions. How serious are these modeling errors at the times and locations where the model predicts the largest sensitivity to ammonia emissions, and what are the implications for interpreting the modeling results?

P5387@L1: “due to chemistry: a shift in chemical” perhaps “due to aerosol thermodynamics: a shift in phase” is more specific.

P5390@L24: This point that nitrogen deposition decreases because near-source *p*-NO_3 decreases seems unusual to me. Near a source, HNO_3 and NH_3 dry deposit quickly relative to $PM_{2.5}$ and should dominate the nitrogen deposition budget. If NH_3 emissions decrease, the fraction of HNO_3 should increase, leading to more near-source oxidized nitrogen dry deposition, not less. It would be helpful to have a table of the total deposition of NH_3, HNO_3, *p*-NO_3, and *p*-NH_4, separated by wet and dry deposition, before and after the emission change, or simply the % change due to the emission change. This would be helpful to compare against the total export across the boundary.

P5394@L6: “predicted decreases in median hourly $PM_{2.5}$ mass of less than or equal to 1 μg m^{-3}” Where is the decrease equal to 1 μg m^{-3}? All of the monitors have median changes less than 1 μg m^{-3}. It would be more helpful to describe when and where the largest changes occur.

P5394@L16: “small but significant decrease” Here it is helpful to be more specific and state that the decrease is a change of 1%.

Technical comments:

Figure 8 is extremely difficult to read. Is it possible to make these plots wider or exclude some of the less relevant data?

References:

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Pinder R. W., A. B. Gilliland, R. L. Dennis. Environmental impact of atmospheric NH 3 emissions under present and future conditions in the eastern United States, *Geophys. Res. Lett.*, 35, L12808, doi:10.1029/2008GL033732, 2008.

Tsimpidi, A.P., V. A. Karydis, S.N. Pandis, Response of Inorganic Fine Particulate Matter to Emission Changes of Sulfur Dioxide and Ammonia: The Eastern United States as a Case Study, *Journal of the Air and Waste Management Association*, Dec. 2007

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 5371, 2009.